

Resolving the interfacial behaviour in complex magnetic nanostructures



OSNS16

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Alternatively...

God made solids, but surfaces were the work of the devil!

Wolfgang Pauli 1900-1958



en.wikipedia.org



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Alternatively...

"There's Plenty of Room at the Bottom: An Invitation to
Enter a New Field of Physics"

RP Feynman 1918-1988



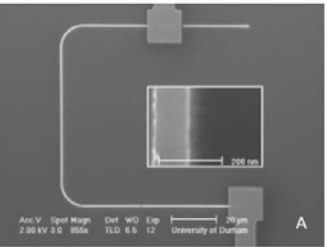
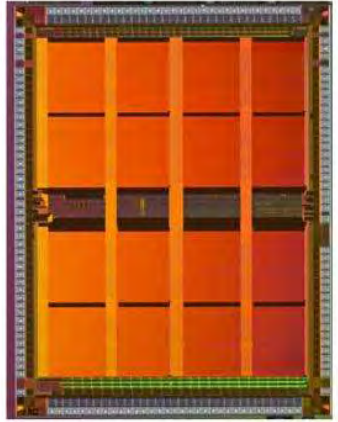
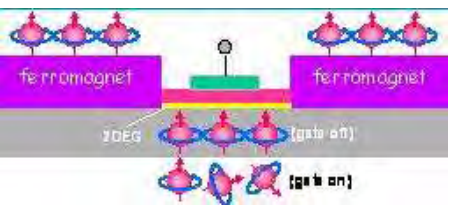
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Grand Challenges: Nanomagnetism



Adapted from S.D. Bader *Rev Mod Phys* **78** (2006)

(Super-)Spintronics

■ Control and manipulation of the spin degree of freedom in the solid state environment

■ Spin

- Transport
- Dynamics
- Relaxation

- Information storage
- Quantum Computing

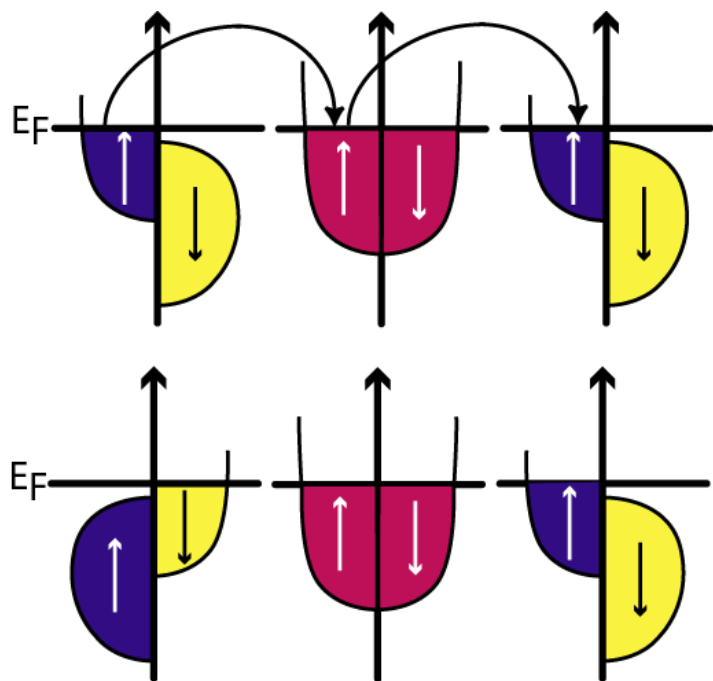
Solid State Physics Lab., Kyushu Univ.

	Charge current	Spin current
Unpolarized current		0
Spin-polarized current		
Fully polarized current		
Pure spin current	0	

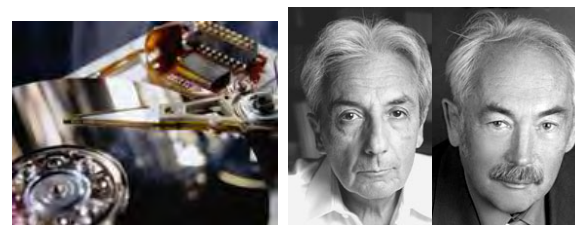
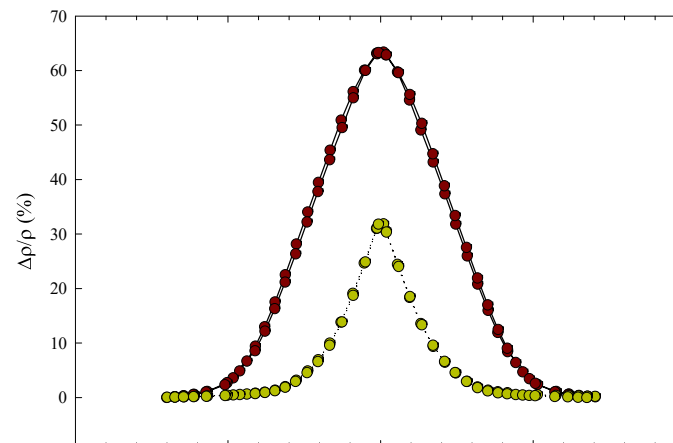


Quantum Well State

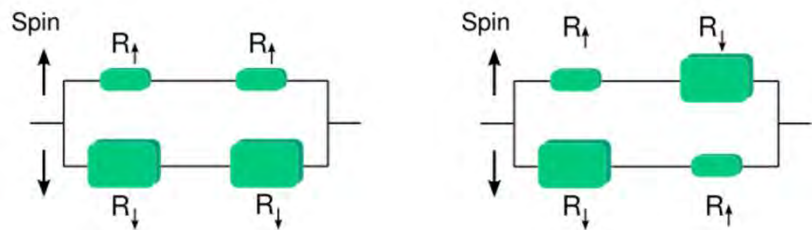
■ Band matching important e.g. Fe/Cr



Co/Cu GMR



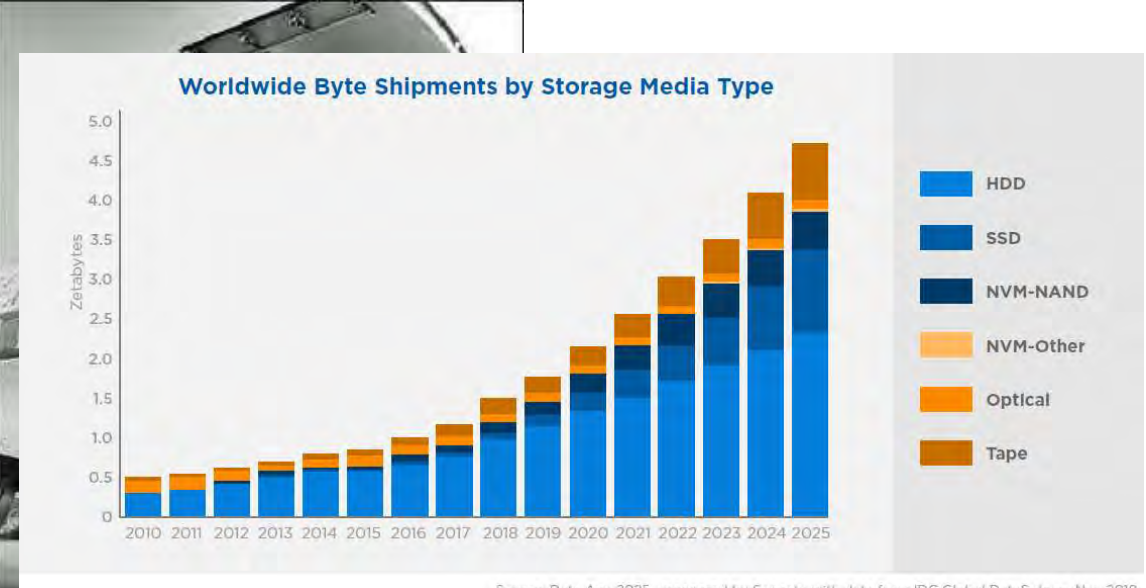
THE
NOBEL
PRIZE



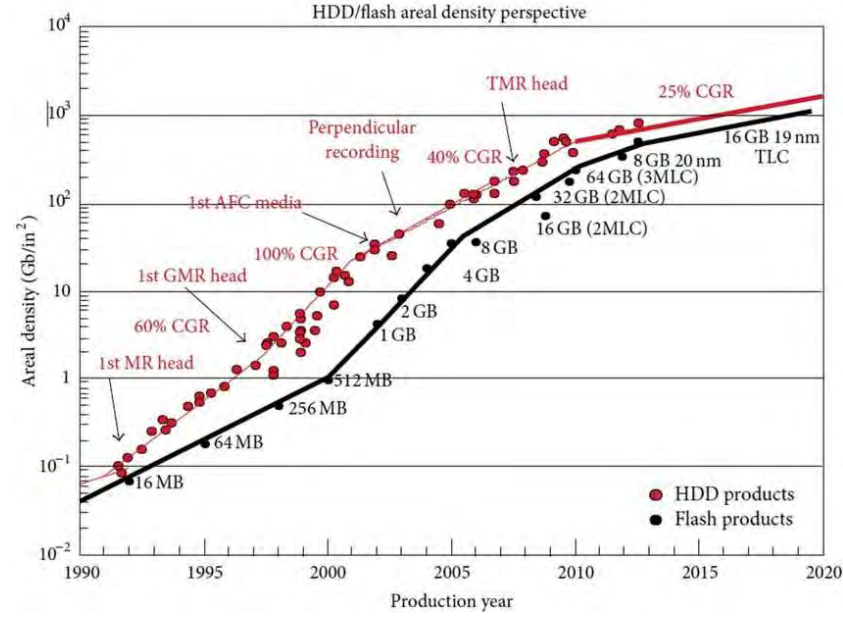
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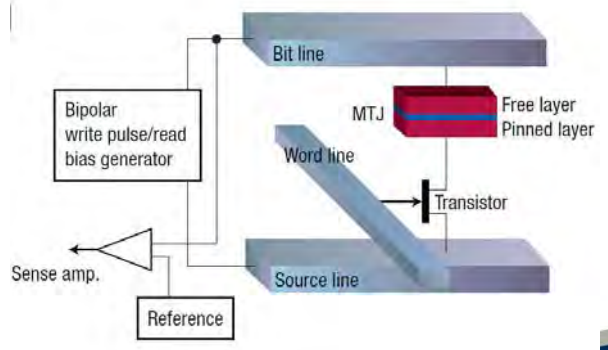
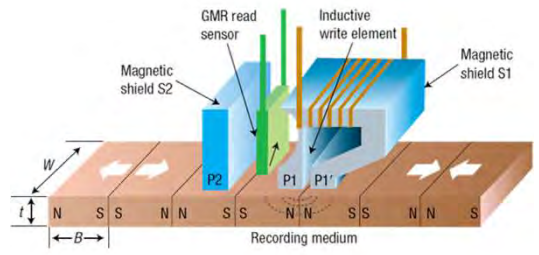
A history of storage/spintronics



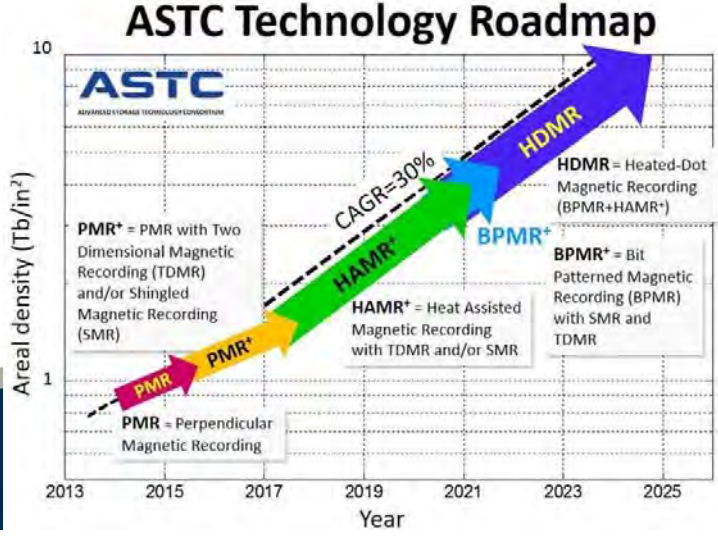
Li Hei Thesis 2016 and refs therein



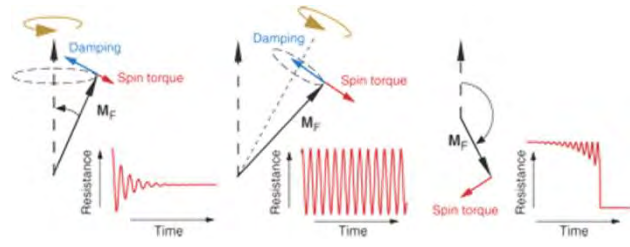
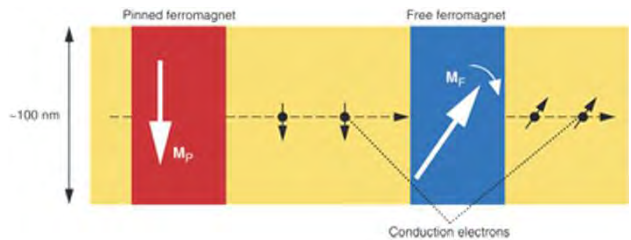
2006
 4Mh MRAM



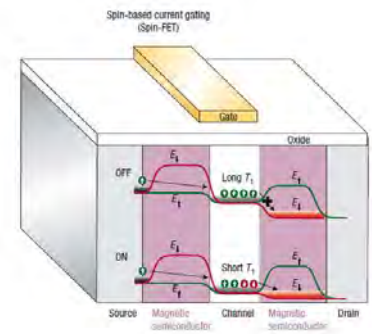
Chappert, Fert, Nguyen van Dau Nat. Matl. 6, 813 (2007)



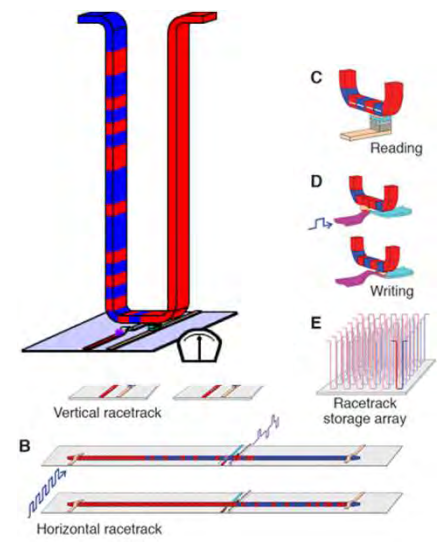
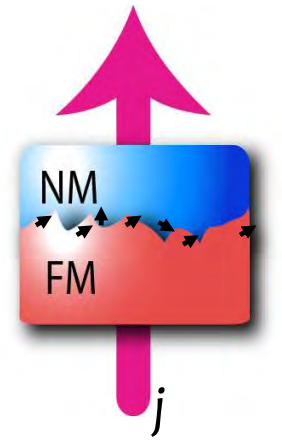
The Importance of interfaces



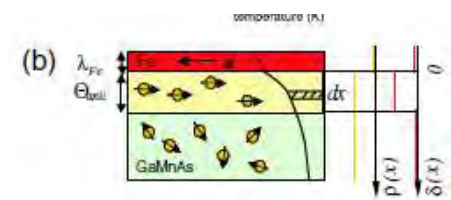
I. N. Krivorotov et al., Science **307**, 228 (2005).



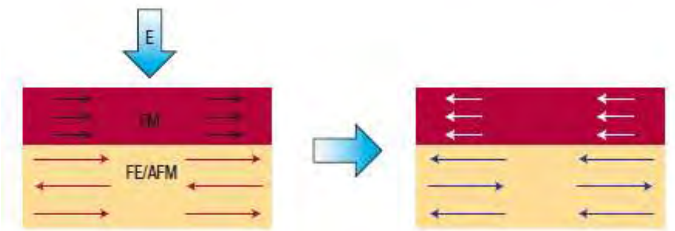
■ Awschalom and Flatté Nat. Phys. **3** 153 (2007)



■ S.S.P.Parkin et al. Science **520** 5873 (2008)

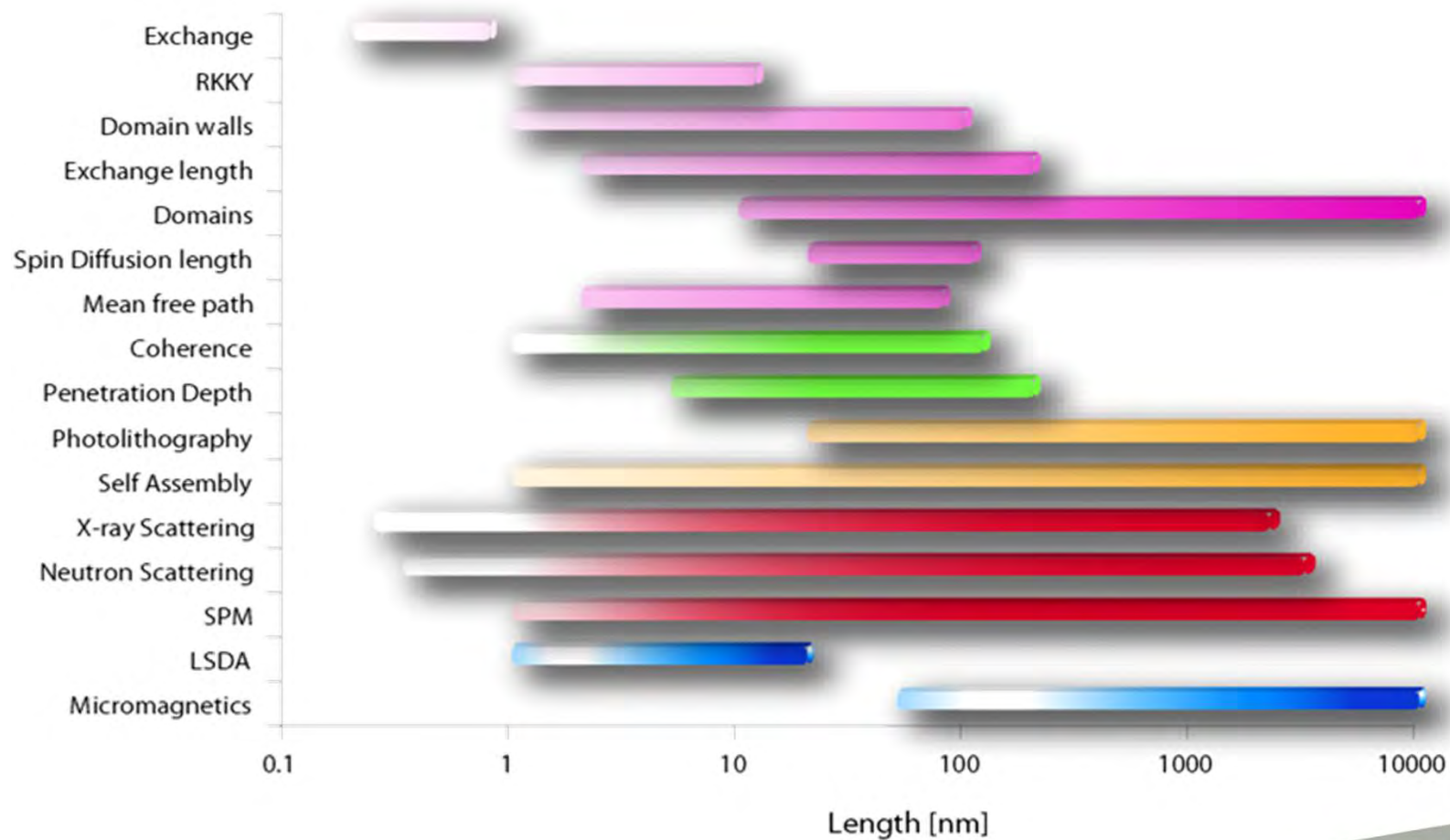


■ Maccherozzi et al. Phys. Rev. Lett. **101**, 267201 (2008)



■ Ramesh and Spaldin Nat. Mat. **6**, 21 (2007)

Relevance of scattering techniques to interfacial effects



Adapted from I.K. Schuller *et al.* J. Magn. Magn.Mater. 200 (1999) 571.



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Nanoscale Electronic Phenomena Research

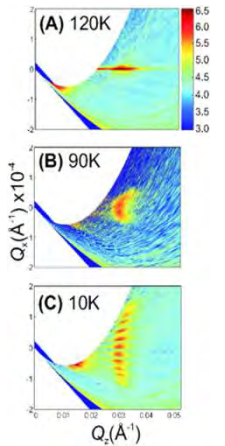
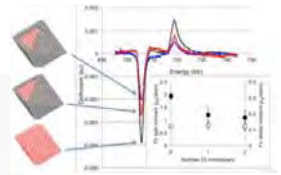
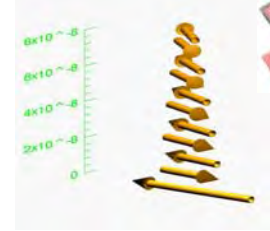
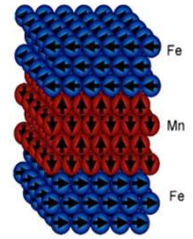
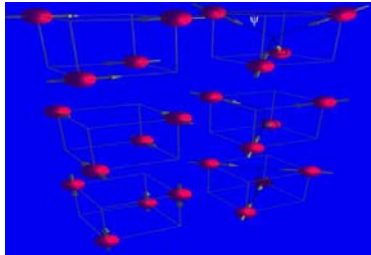
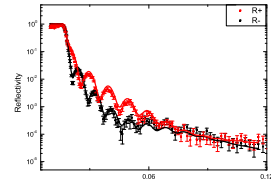
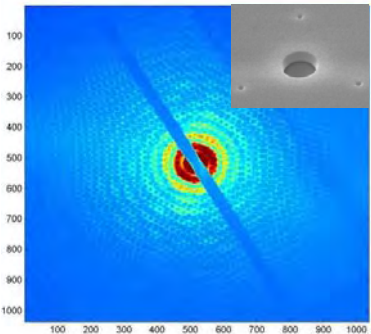
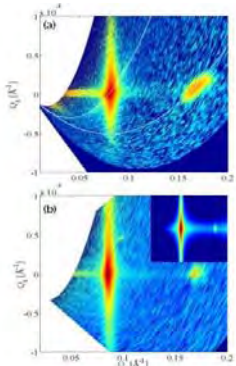
Heusler, DMS

Surface Magnetism

Exchange Bias

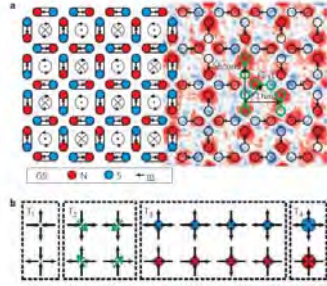
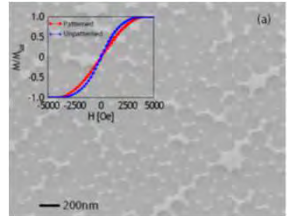
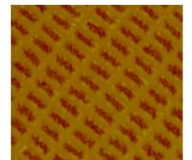
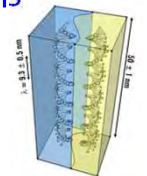
- Conventional EXB
- Synthetic EXB
- Frozen magnetism

Domain Structures

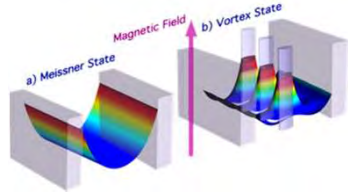
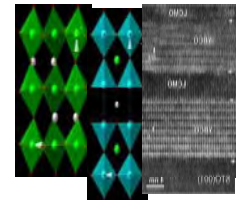


- To understand and control spin and interfacial phenomena
 - Probe length scale (<1nm to >1000nm)
 - Vector Magnetometry $\sim <0.1\mu_B$ per f.u.
- Frustration

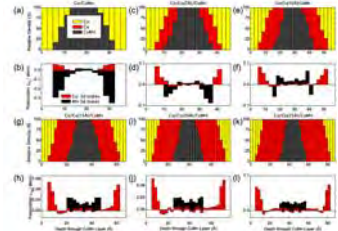
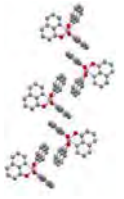
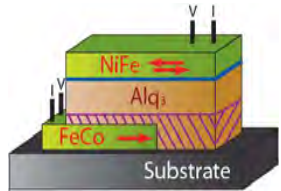
Helimagnetism-Skyrmions



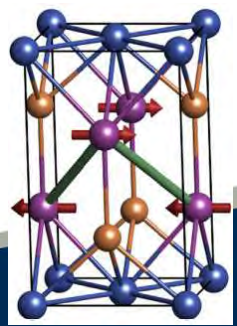
Singlet-Triplet Superconductivity



Organic Spintronics



AF Semiconductors



Motivation & Outline

■ Motivation

- *Unique and quantitative* description of complex electronic materials on the microscopic lengthscale
- Relating the functional properties of materials to their atomic and nanoscale structure

■ Introduction to Polarised Neutron Reflectivity (PNR)

■ Ferrimagnetic insulators for spintronics/magnonics

- *Understand interfacial behaviour in spin-current, magnonic systems*
- *Understand some of the low-T anomalies in thin ferrimagnetic insulator films*
- *Characterise the spin axis on FI/AFI systems*

■ Chiral Magnetism

- *Control of helical structures*
- *Study of DMI in thin films*

■ Magnetocaloric Material

■ Towards Super-Spintronics

■ Small Angle Scattering

■ Summary & Conclusions



Acknowledgements

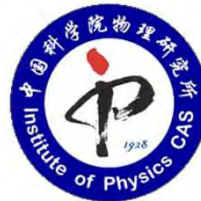


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- B. J. Hickey
- A. Mitra
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- C.S. Spencer



- S. Singh
- S. Basu



- T. Zhu



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- J.F.K. Cooper
- D Alba Venero
- A. Caruana



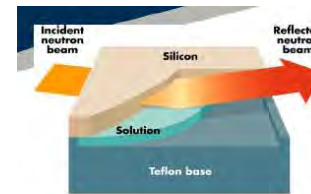
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Recall Webster Talk Last Week

NEUTRON REFLECTIVITY

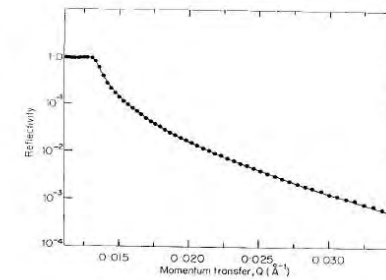


(simple II

Within Born Approximatio
Reflectivity is given as,

$$Q = k_1 - k_2 = 4\pi \sin \theta / \lambda$$

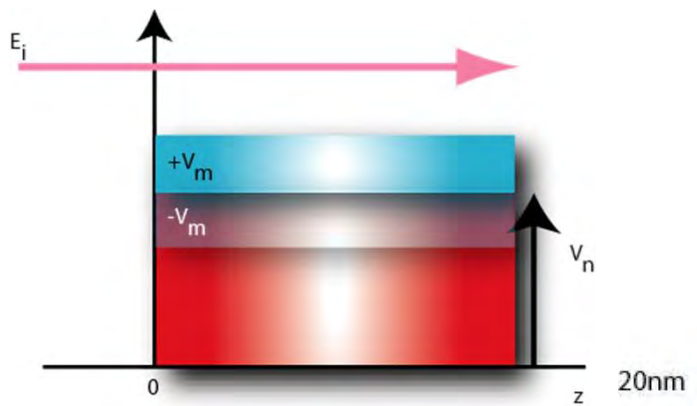
Reflectivity from a simple single interface is then g



R

↓

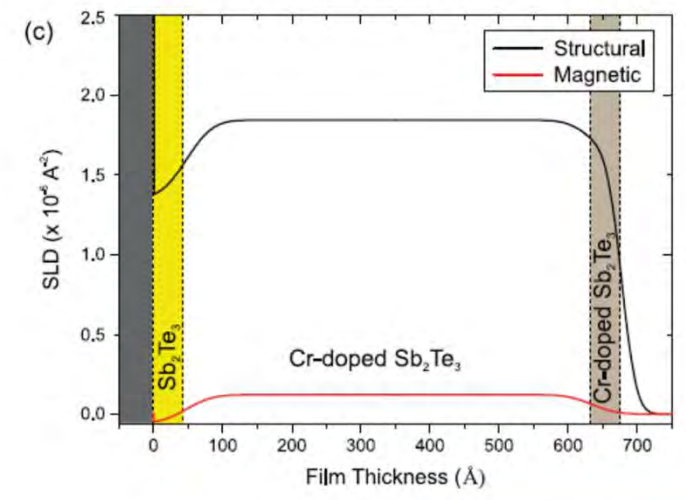
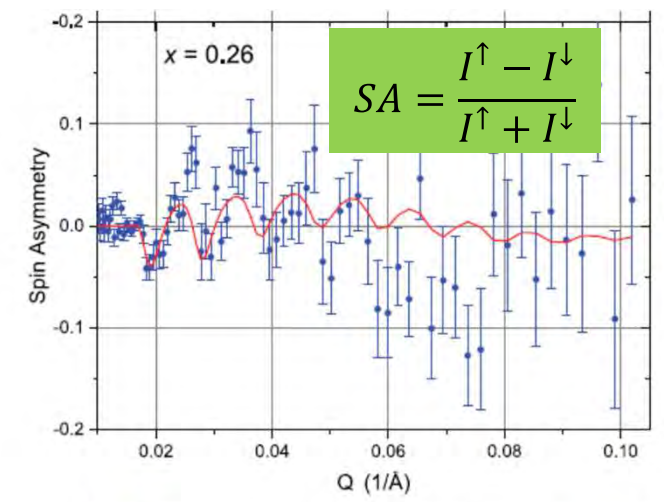
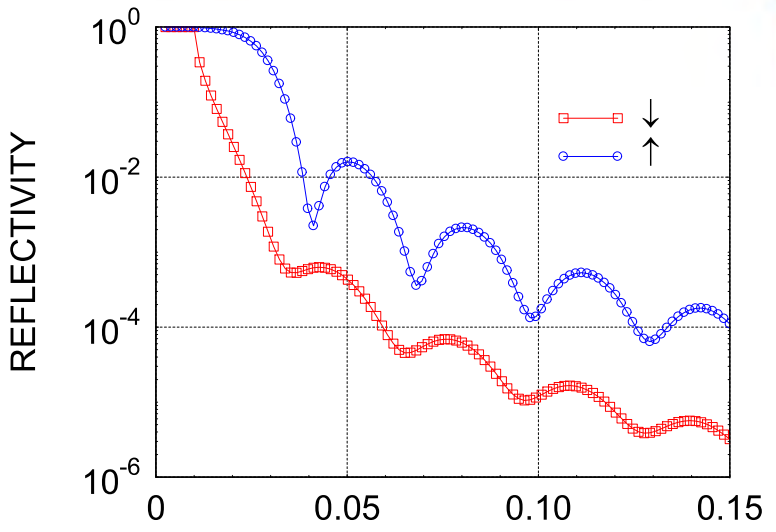
PNR from a single layer



$$V = V_n + V_m$$

$$V = \frac{\hbar^2}{2\pi m} N(b \pm p)$$

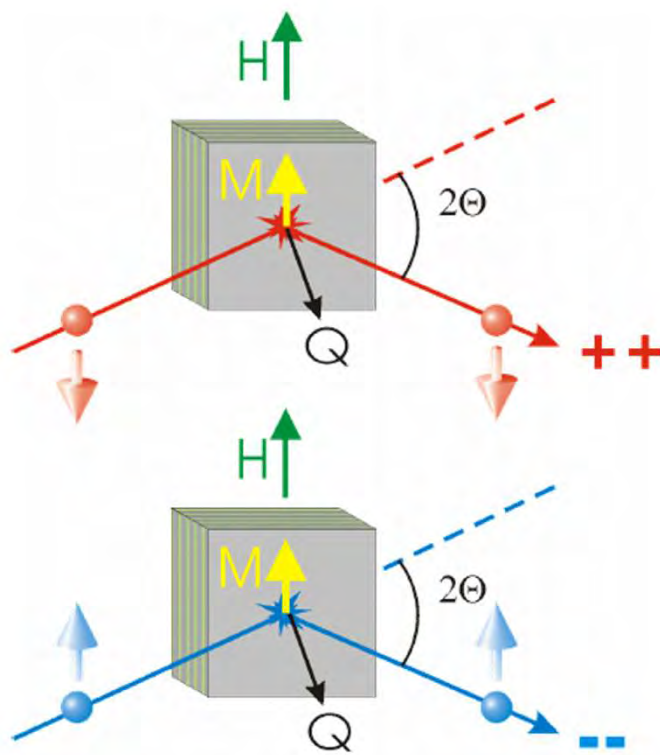
$$p = (2.695 \times 10^{-4} / \mu_B) |\vec{\mu}_i|$$



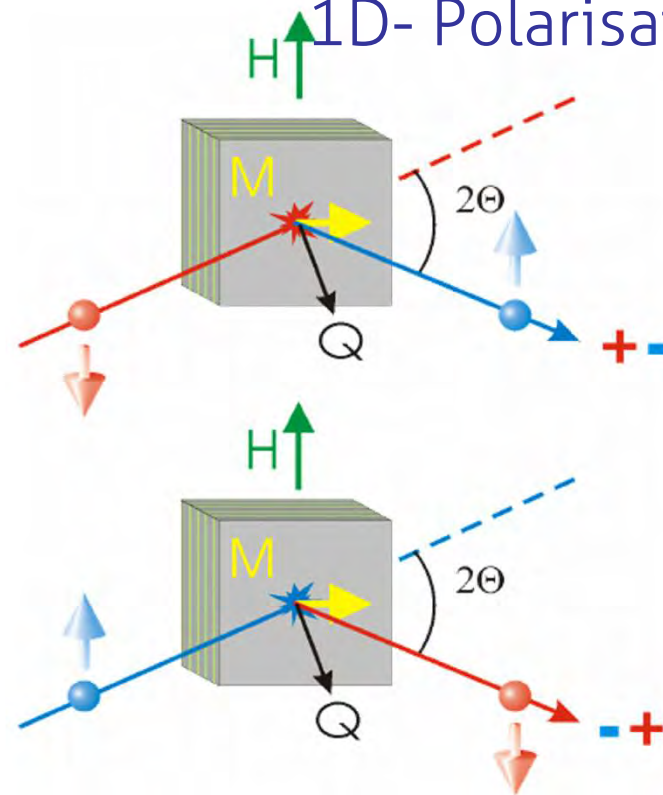
$Q_z [\text{Å}^{-1}]$

Collins-McIntyre, L. J. *et al.*
Europhysics Lett. **115**, 27006 (2016)

1D- Polarisation Analysis



Non spin flip
 ++ measures $b + M_z$
 -- measures $b - M_z$



Spin flip
 + - measures $M_x + iM_y$
 - + measures $M_x - iM_y$

By fitting all components the direction and strength of the magnetic moment can be measured as a function of depth



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PoIREF BEAMLINE

- TOF wavelength band 1Å – 16Å
- Vertical and horizontal geometry
- Non-polarised, polarised and polarisation analysis modes
- Sample point goniometer capable of moving 1000kg
 - GMW Magnet ($\pm 1T$).
 - Cryostat (2.5K -300K), (sub 1K fridge) ad hoc in-situ transport.
 - Vacuum furnace (300K – 800K).
 - PNR/PA Polarised modes.
 - Various soft matter setups
 - H loading
- Experimental Setups:



Generating 'Pure' Spin currents: Ferrimagnetic Insulators

Understanding the interfacial behaviour

'As spin pumping in heterostructures requires the transfer of spin information across an interface, the role of interface quality must be understood thoroughly, yet it remains unclear.'

Gray, M. T. *et al.* Phys. Rev. Appl. **9**, 064039 (2018)



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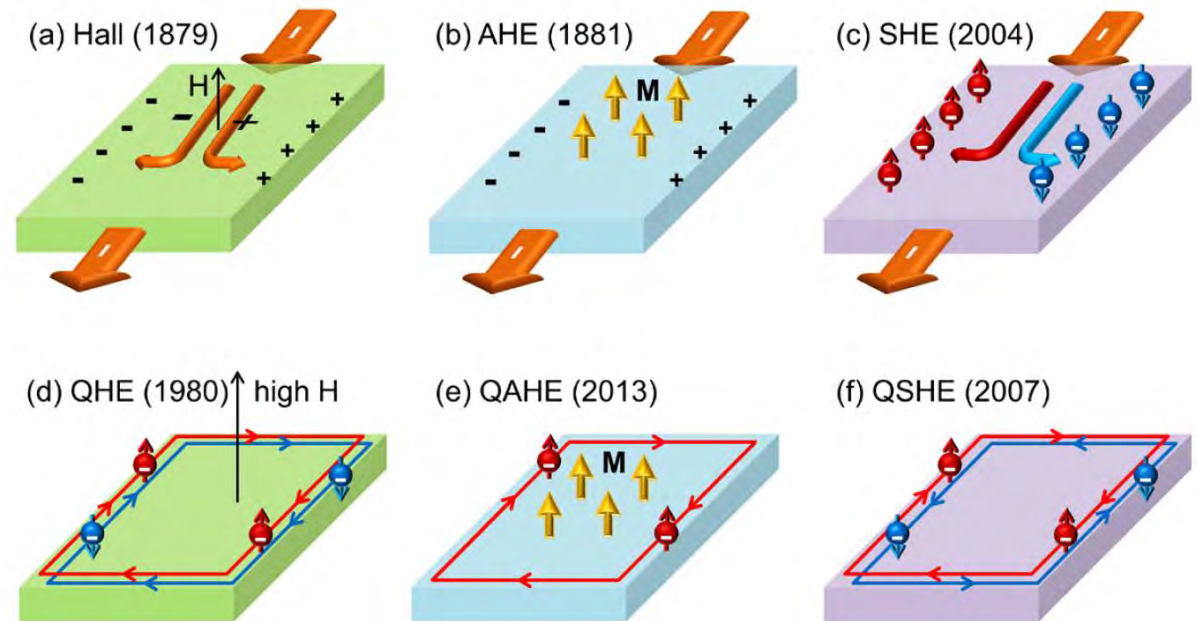
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Meet the Hall family..



'On a new action of the Magnet on Electric Currents'
American Journal of Mathematics vol 2, 1879, p.287-292

- For a century only the two effects
- Extrinsic vs. intrinsic
 - SOC impurities
 - ◆ E.g. through spin injection
- Ideal for sensor technologies to low-power technologies
 - Spin transistors, spin logic, and spin quantum computing



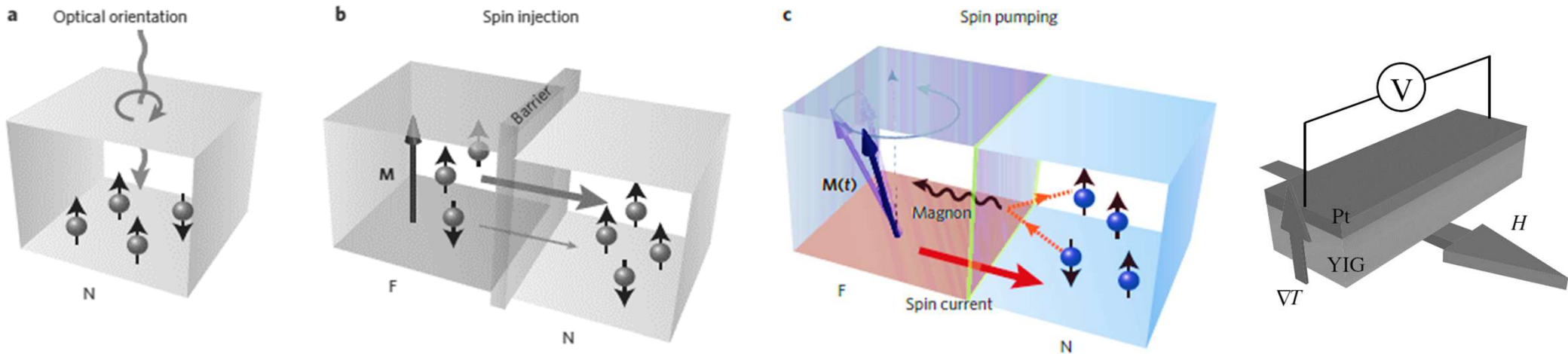
Chang, C.-Z. & Li, M. Quantum anomalous Hall effect in time-reversal-symmetry breaking topological insulators. J. Phys. Condens. Matter **28**, 123002 (2016).



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Generate a spin imbalance

■ Various mechanisms



Qiu, Z., Hou, D., Uchida, K. & Saitoh, E. Influence of interface condition on spin-Seebeck effects. *J. Phys. D: Appl. Phys.* **48**, 164013 (2015).

Žutić, I. & Dery, H. Spintronics: Taming spin currents. *Nat. Mater.* **10**, 647 (2011)

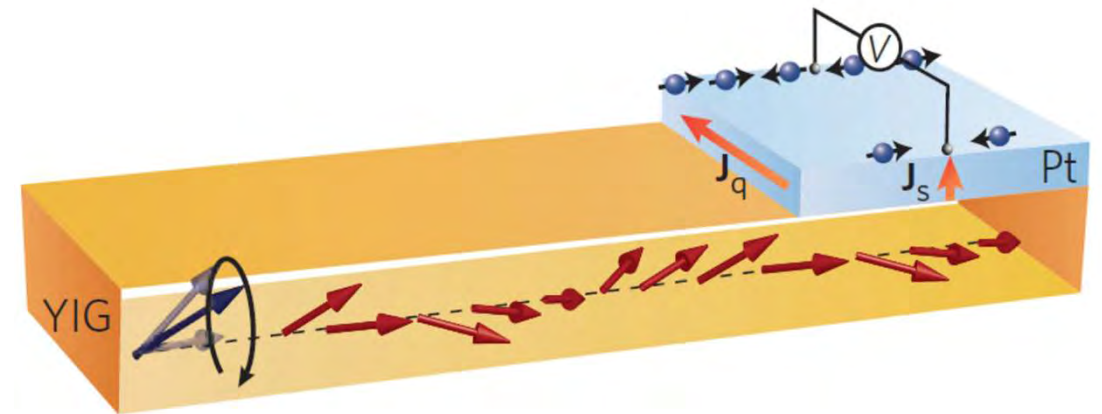


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Inverse Spin Hall effect

- Exploit the SOC
 - Spin and orbital motion are coupled
- 🙄 ■ SOC limits spin diffusion length
 - Channel for spin relaxation
- SOC makes effective sink for the angular momentum
- Applying a field increases the number of magnons and the lattice acts as a source of spin current
- Use YIG as a source of spin current
- Pt as a spin converter



Žutić, I. & Dery, H. Spintronics: Taming spin currents. *Nat. Mater.* **10**, 647 (2011)



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YIG: $Y_3Fe_5O_{12}$

■ YIG ferrimagnetic insulator

■ Ferrimagnetic insulator

- ◆ Bandgap 2.58eV
- ◆ $T_c = 560K$

■ Transparent above 600nm

■ Low absorption in IR

■ Very low damping

■ Small linewidth for esr

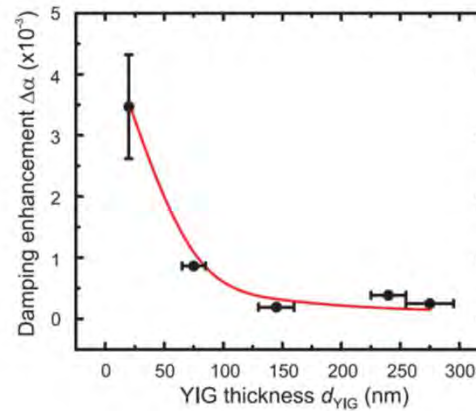
■ Long spin wave decay length/ magnon damping

■ Ideal for:

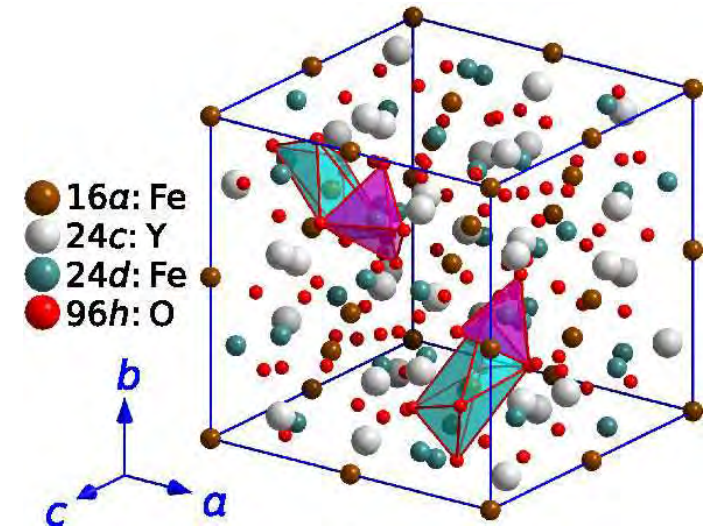
■ optical and magneto-optical applications,

■ microwave filters

■ Spin Seebeck/Peltier applications



Jungfleisch, M. B. *et al.* Thickness and power dependence of the spin-pumping effect in YIG/Pt *Phys. Rev. B* **91**, 134407 (2015).



A. Kreisel, Goethe Univ.

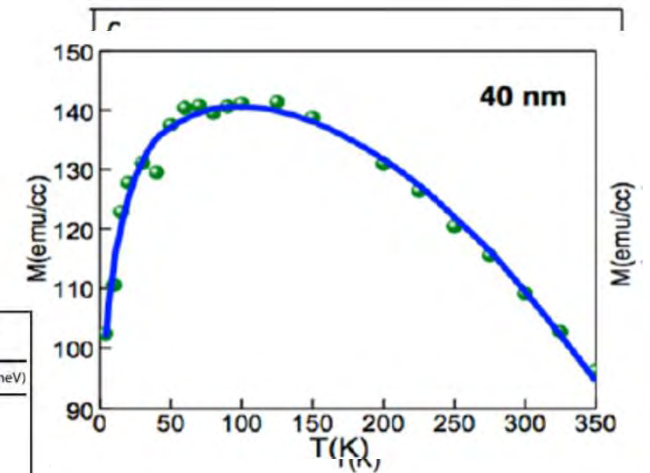


Table 1. Fitted exchange parameters for YIG and their statistical uncertainties

	This work (meV)	Ref. 3 (meV)	Ref. 23 (meV)	Ref. 21 (meV)
J_1	6.8(2)	6.87	4.774	6.4
J_2	0.52(4)	2.3	0.308	0.9
J_{3a}	0.0(1)	0.65	0.144	0
J_{3b}	1.1(3)	0.65	0.144	0
J_4	-0.07(2)	-	0.326	0.46
J_5	0.47(8)	-	0.358	0.28
J_6	-0.09(5)	-	0.008	1.5

The exchange interactions are defined in Fig. 1, and are per pair of spins

- Princep, A. J. *et al.* The full magnon spectrum of yttrium iron garnet. *npj Quantum Mater.* **2**, 63 (2017).

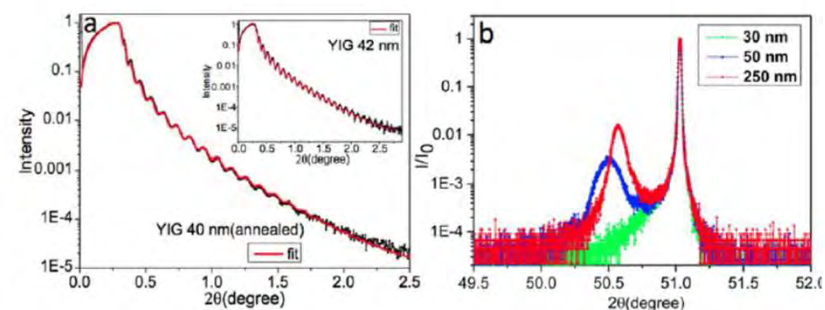
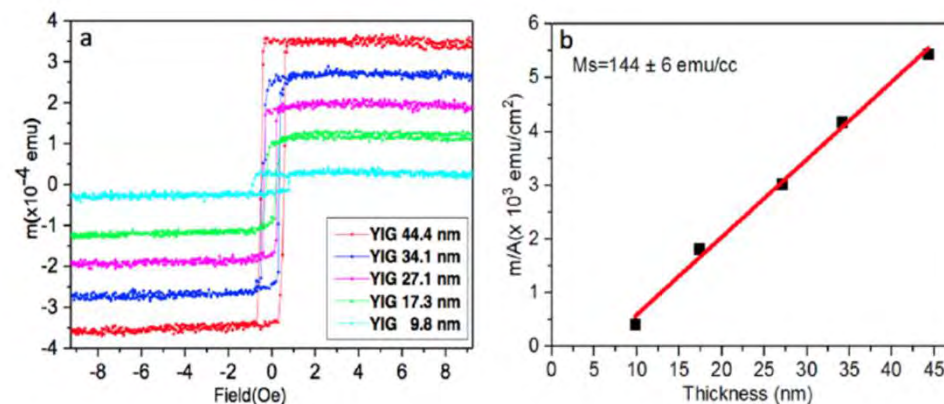
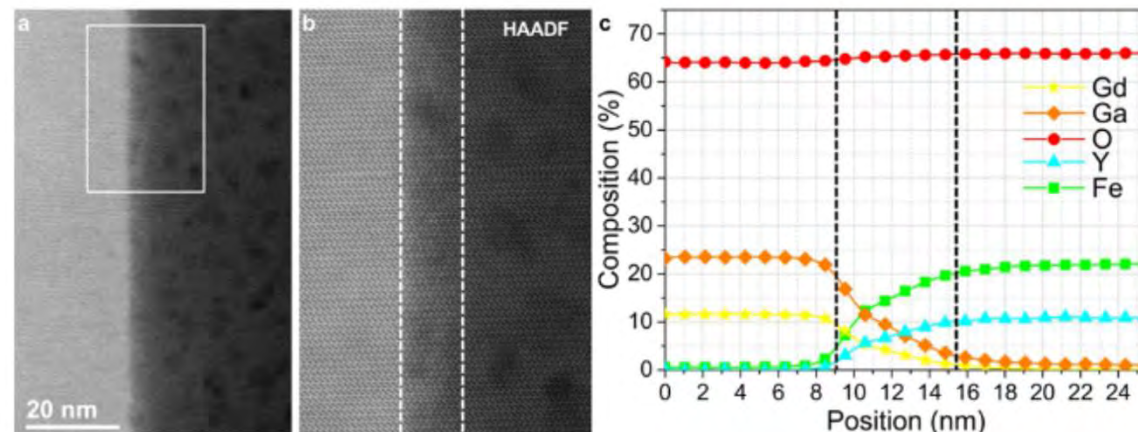


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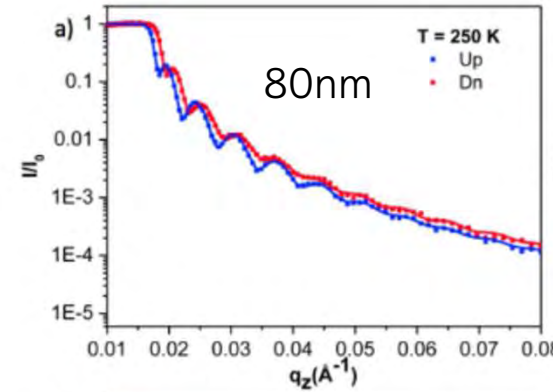
Thin film YIG

- Numerous Growth techniques
- RF sputtered followed by anneal 850c for 2 hrs
- Grow on GGG ($Gd_3Ga_5O_{12}$)
- 0.06% lattice mismatch
- SuperSTEM
 - Abberation corrected
 - HAADF
 - 6nm diffusion
 - Diffusion co-efficient $1 \times 10^{-17} \text{cm}^2 \text{s}^{-1}$ (agrees with bulk)



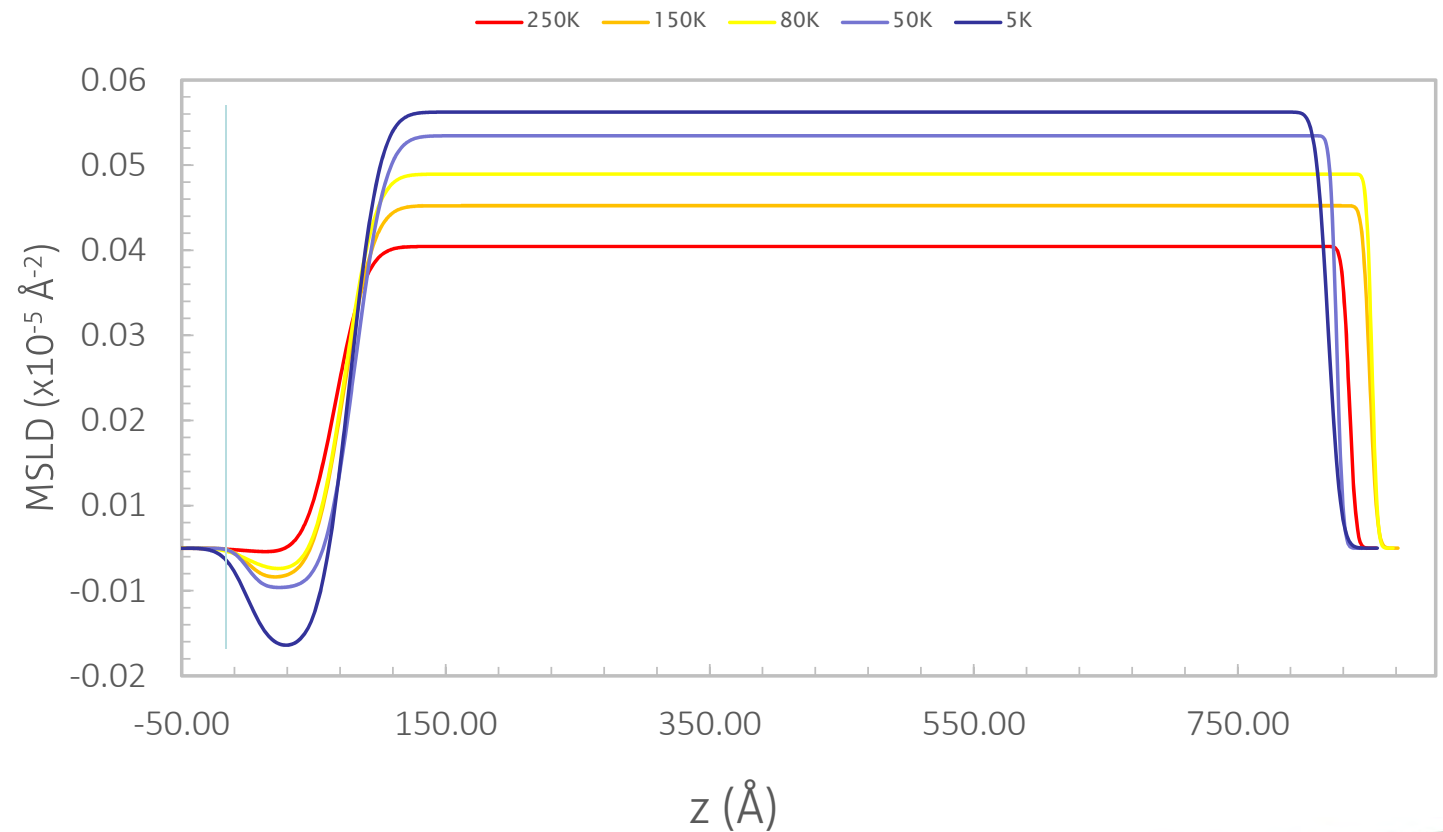
PNR Data

- Analysed within a simple 2-layer model
 - 2 layer model
 - ◆ Pristine YIG
 - ◆ Gd doped YIG
 - ◆ $3.8\mu_B/\text{u.c.}$
 - Gd 6-7nm diffusion
 - Excellent agreement with x-ray and SuperSTEM
 - Gd absorption cross-section enhances sensitivity
 - Gd Iron Garnet: room temperature compensation



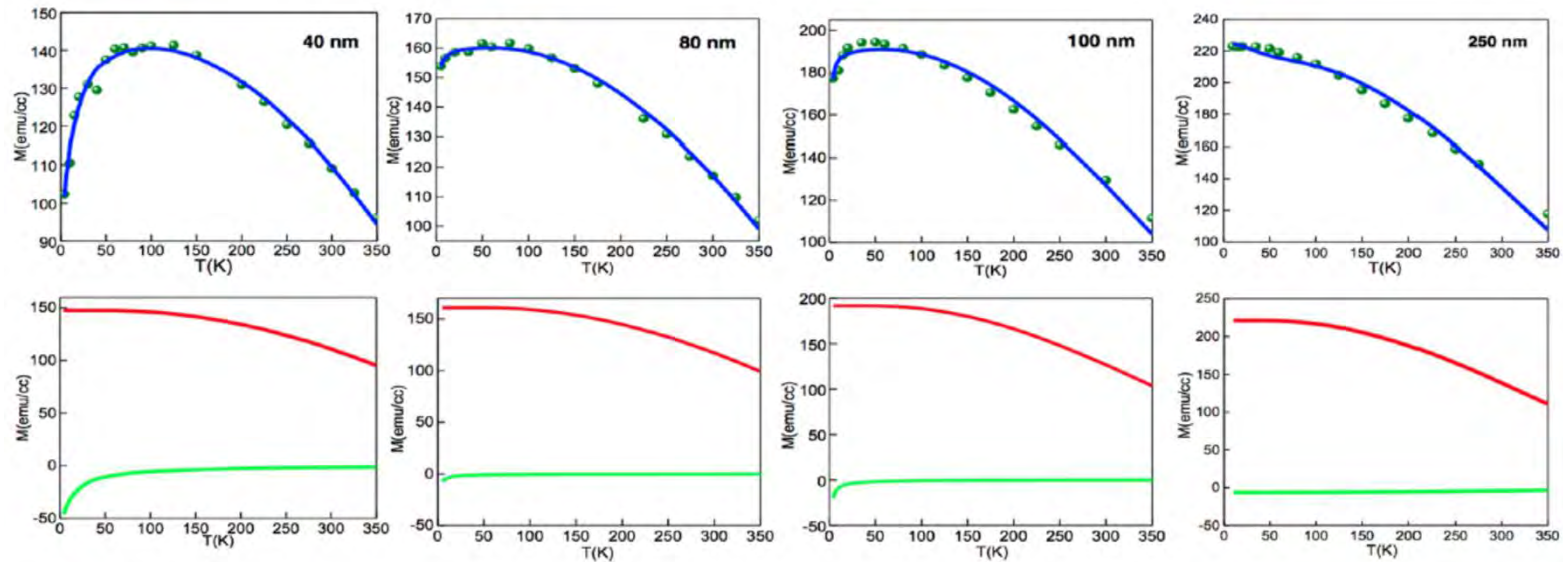
Temperature Dependence

- Gd Iron Garnet: room temperature compensation
- $M(T)$ YIG follows Bloch law
- Antiparallel moment develops at low temperature near to interface
- Gd substitutes on Y site
- Unusual magnetisation temperature dependence from Gd diffusion
- Effect on low-T FMR linewidth
- ISHE correlates with interface quality
- Probably not the full story...

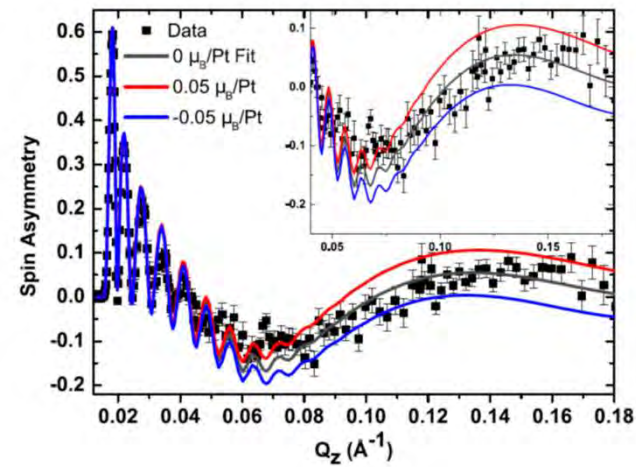
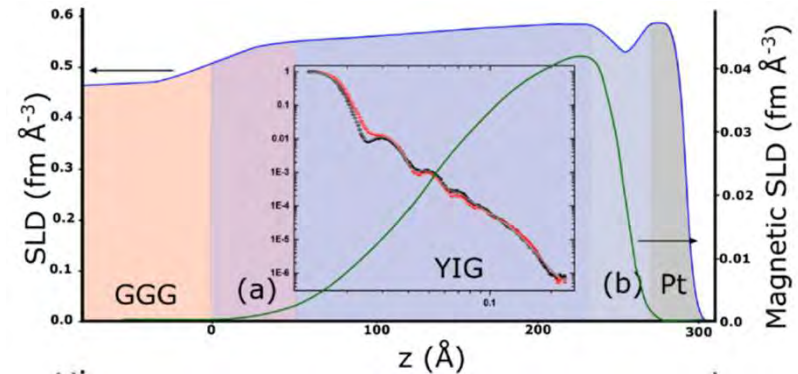
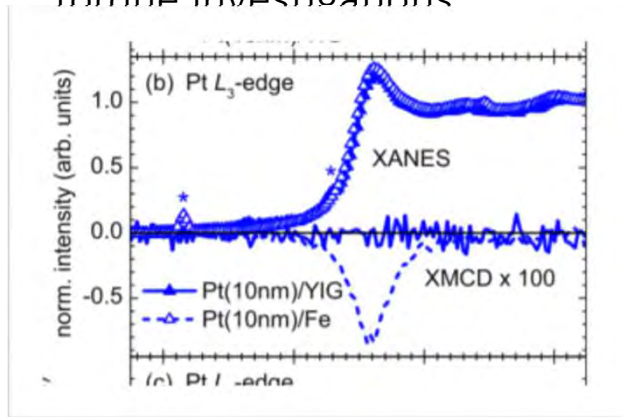


■ Mean Field Model

$$■ M(T) = M_y B(J_y, z_y) + M_g B(J_g, z_g)$$



- No observable interfacial moment
- XMCD: $0.003\mu_B$ averaged
- Effect on low-T FMR linewidth
- Reduced Spin Seebeck
- Possibilities to lower growth temperature to limit diffusion
- ISHE correlates with interface quality
- Relevance to spin pumping, spin torque investigations



S. Gebrägs *et al.* Appl. Phys. Lett. **101**, 262407 (2012);



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ISIS

MECHANISM FOR THE -VE SPIN HALL MR



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Spin Hall Magnetoresistance (SMR)

SHE

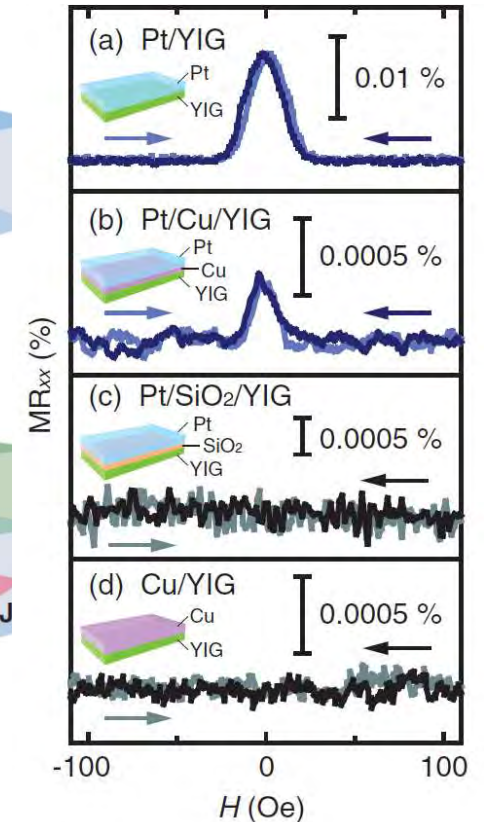
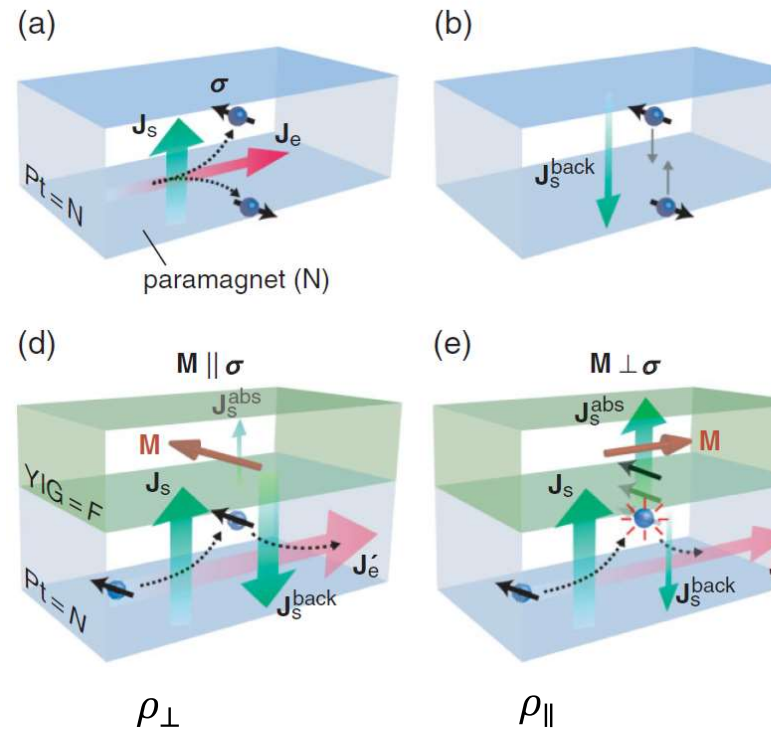
- Conversion of electric current into a transverse spin current
- Generates spin current and accumulation

How can we control/observe this?

- Ferr(i)omagnetic insulator
- Interfacial spin mixing
- Pt film resistance depends on the YIG orientation

SMR

- $\rho_{\parallel} > \rho_{\perp}$



Nakayama, H. *et al.* Spin Hall Magnetoresistance Induced by a Nonequilibrium Proximity Effect. *Phys. Rev. Lett.* **110**, 1–5 (2013).

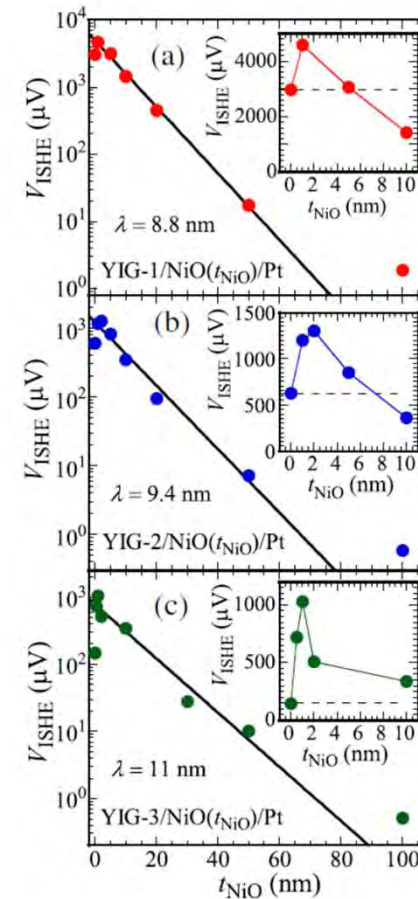


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SMR

- Experimentally verified in:
 - Pt/YIG, Ta/YIG, W/CoFeB
- Pt(4)/NiO(1-2)/YIG(10)/GGG
- Spin current enhanced if an AF layer is inserted (NiO)
 - High spin transfer efficiency
 - Spin current mediated by magnons
- Expect +ve SMR
 - Characteristic temperature (below $T_{\text{Néel}}$)
 - Changes sign at low temperature
 - ◆ Spin flip scattering → Inversion of $J_{\text{e,add}}$ *PRL* **118**, 067202 (2017)
 - ◆ Spin flop coupling *PRL* **118**, 147202 (2017)
 - Spin axis of NiO $\perp M_{\text{YIG}}$



Wang, H., Du, C., Hammel, P. C. & Yang, F. Antiferromagnonic Spin Transport from YIG into NiO *Phys. Rev. Lett.* **113**, 097202 (2014).



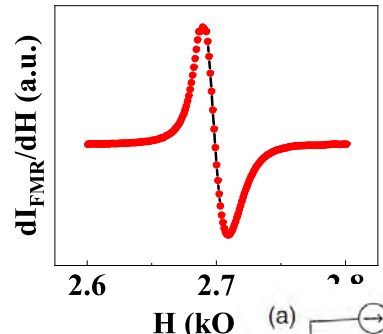
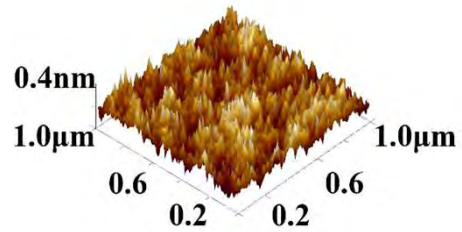
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GGG/YIG(15)/NiO(2)/Pt(4nm)

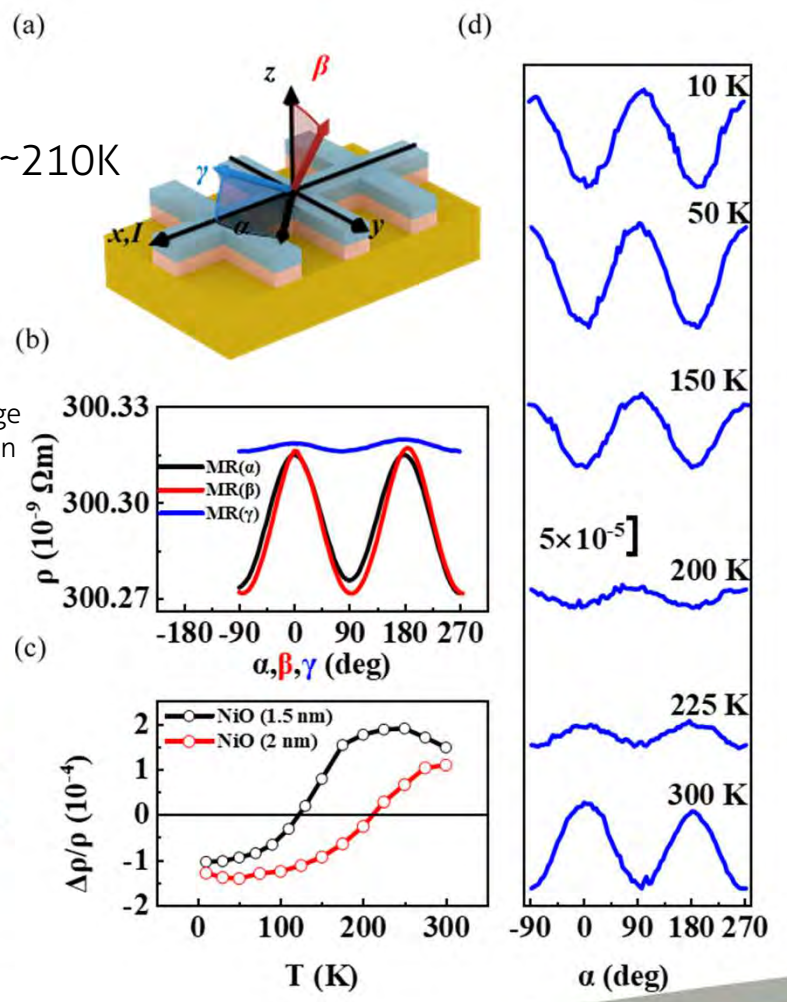
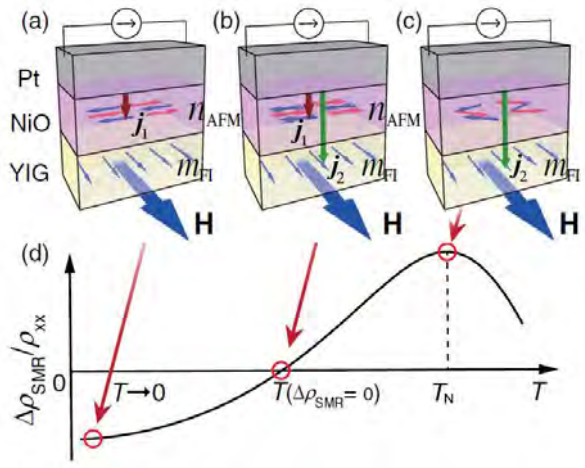
- PLD
- RT, 9.7798GHz
- Resonance field comparable to bulk
- $M = 145 \text{ emu/cm}^3$

- +ve SMR@RT
- Change in sign at ~210K



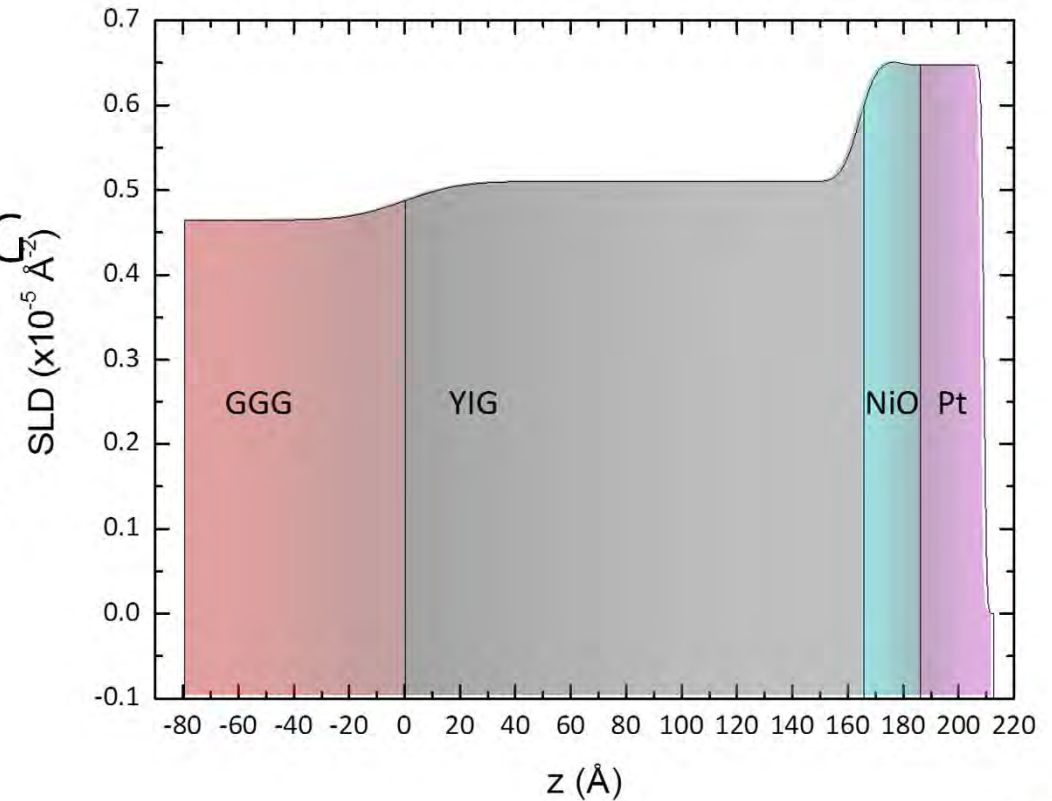
Hou, D. *et al.* Tunable Sign Change of Spin Hall Magnetoresistance in Pt/NiO/YIG Structures. *Phys. Rev. Lett.* **118**, 147202 (2017).

- Expect +ve SMR
 - Characteristic temperature (below $T_{\text{Néel}}$)
 - Changes sign at low temperature
 - ◆ Spin flip scattering \rightarrow Inversion of J_{add} *PRL* **118**, 067202 (2017)
 - ◆ Spin flop coupling *PRL* **118**, 147202 (2017)
 - Spin axis of NiO $\perp M_{\text{YIG}}$

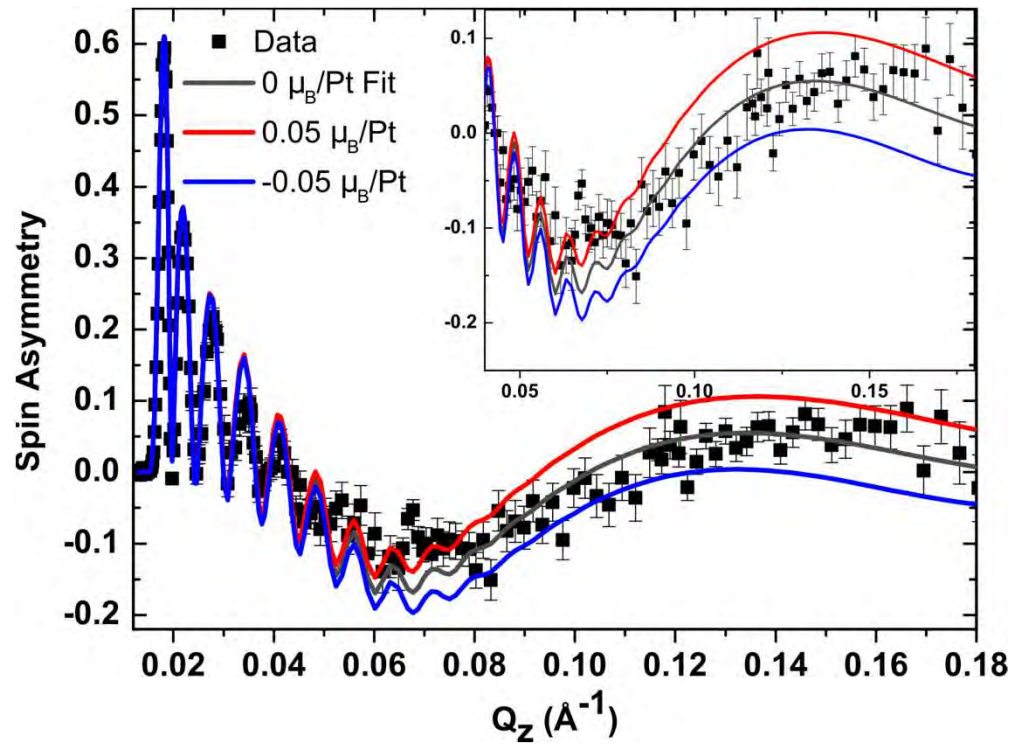


Structural Profile

- Structural profile from PNR
- Pt (4 nm)/NiO (2 nm)/YIG/GGG
- Agreement with XRR & AFM
- Evidence for uncompensated moment?



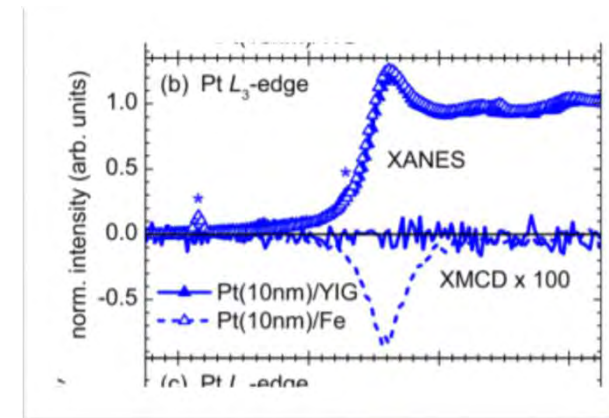
Induced Pt moment (proximity/accumulation)



Spin accumulation at the interfaces

- ◆ Sensitivity to $<0.05\mu_B/\text{Pt}$
- ◆ Less than $0.02\mu_B/\text{Pt}$ (within 1σ)
- ◆ XMCD: $0.003\mu_B$ averaged

S. Gebrägs *et al.* Appl. Phys. Lett. **101**, 262407 (2012);

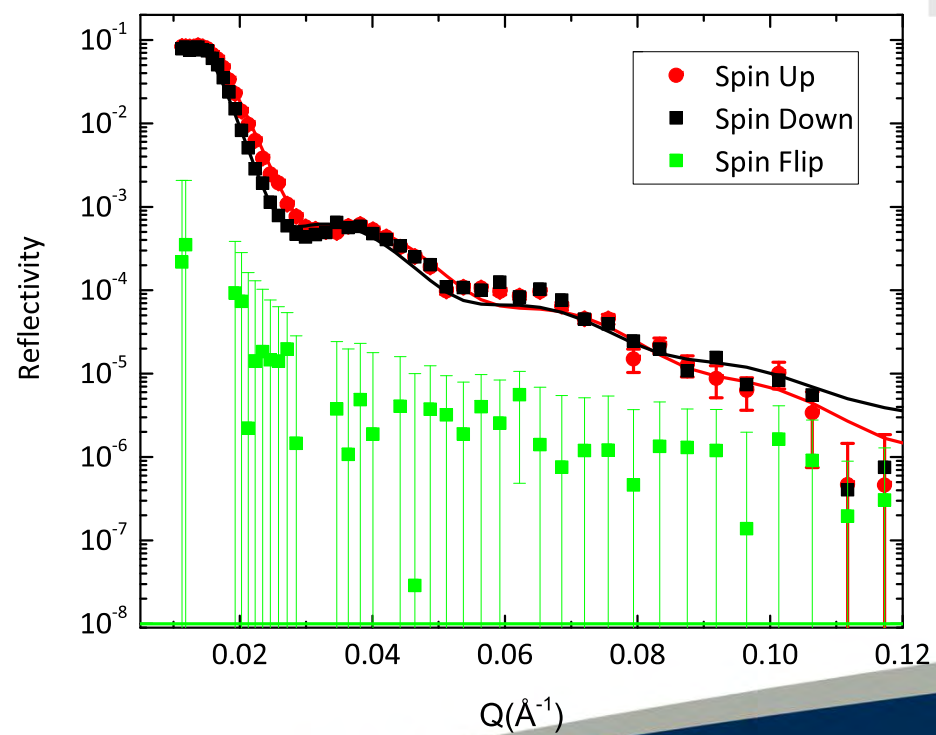
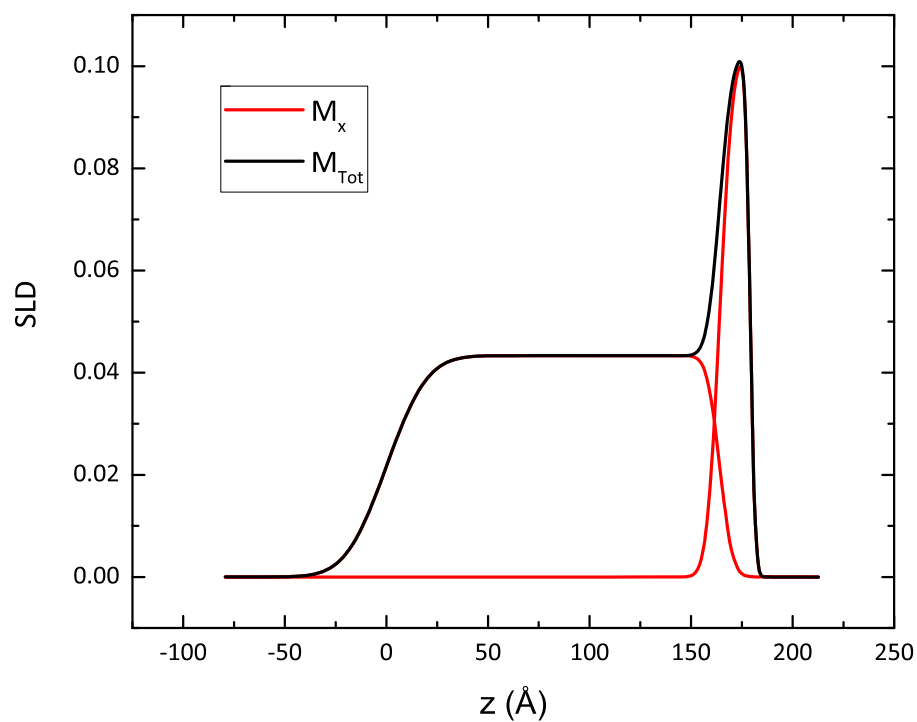
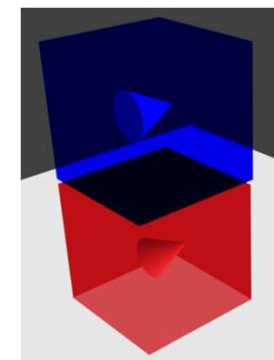


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Parallel Alignment

■ Does not describe SF data

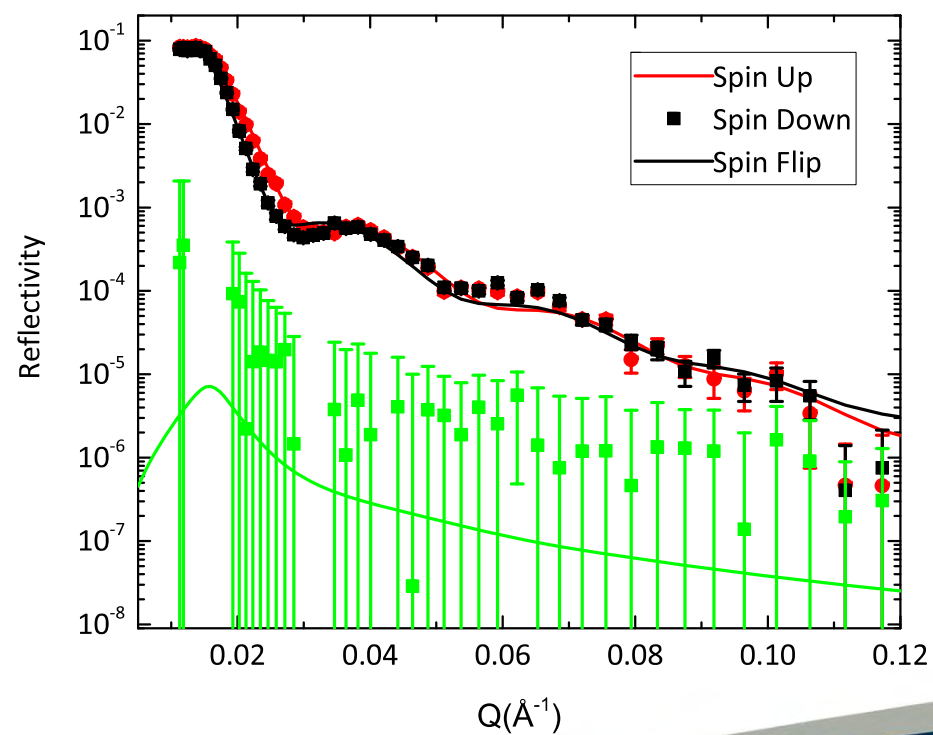
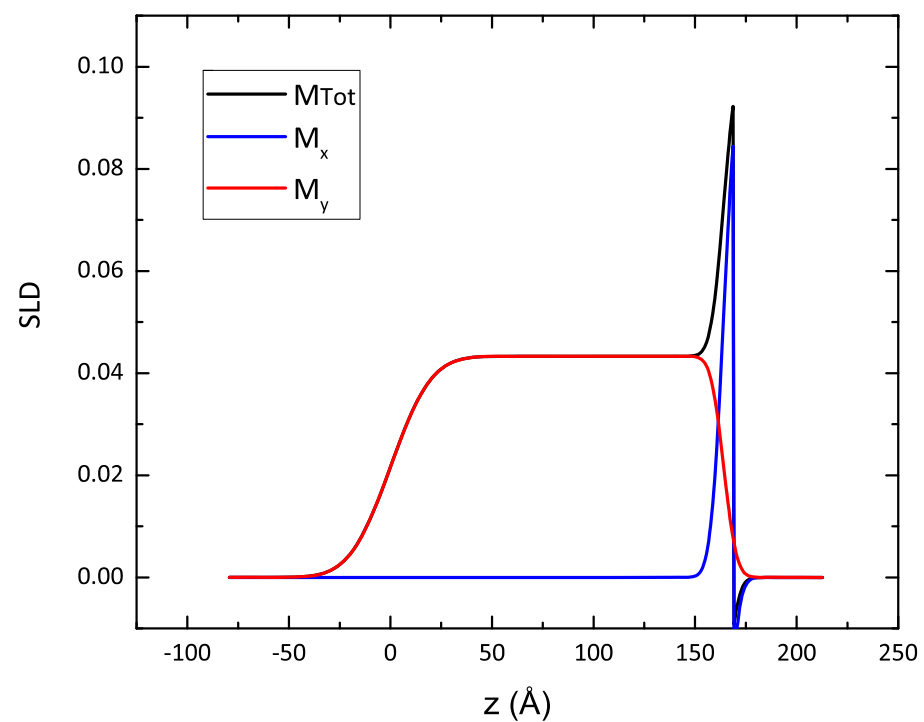
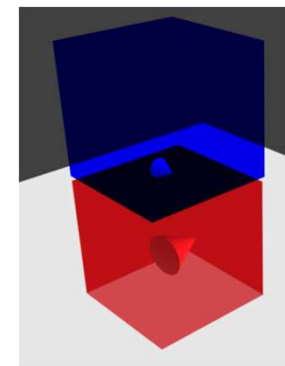


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Interfacial NiO layer

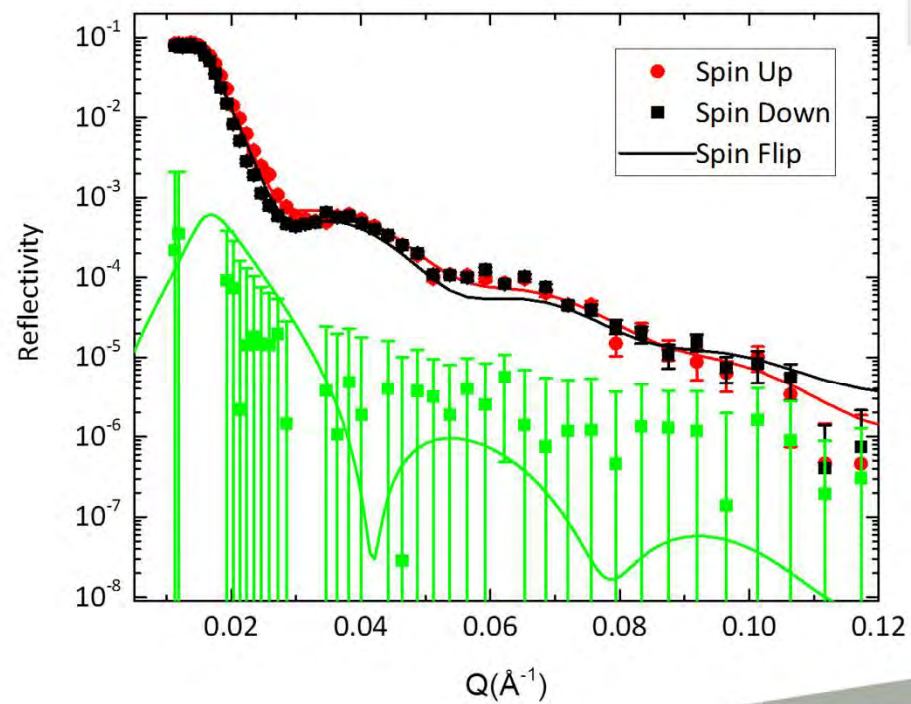
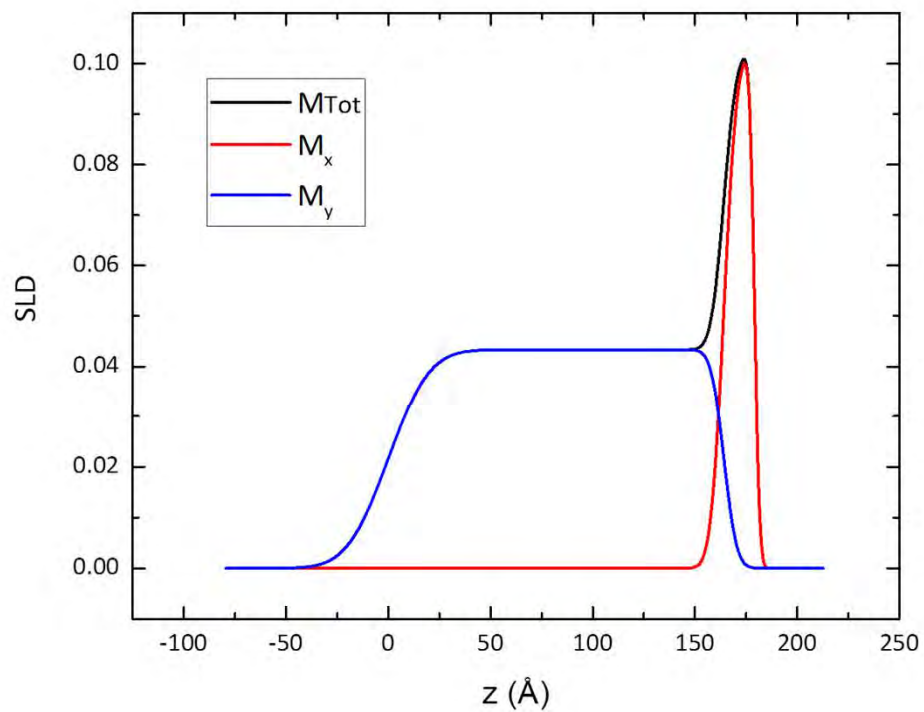
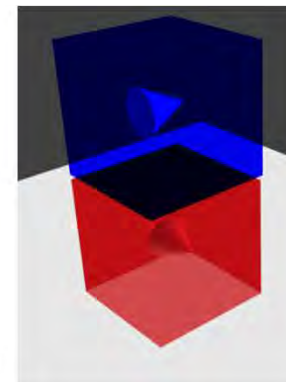
■ Promising...



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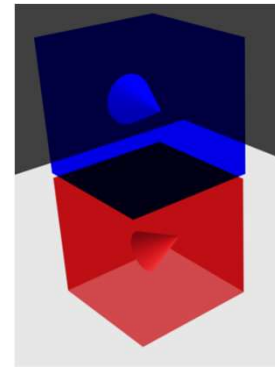
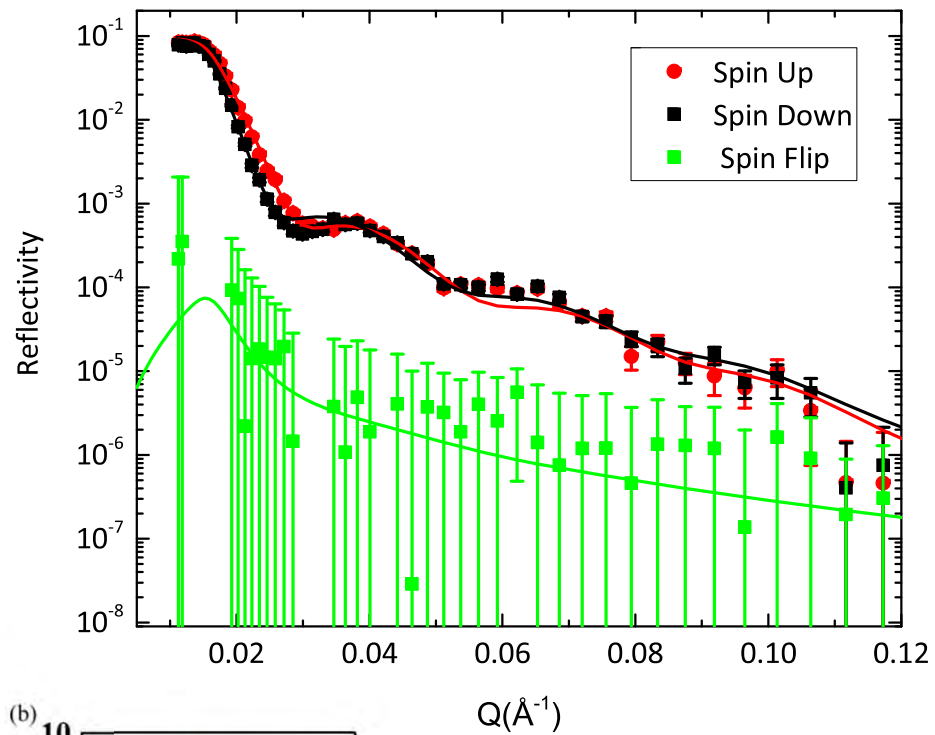
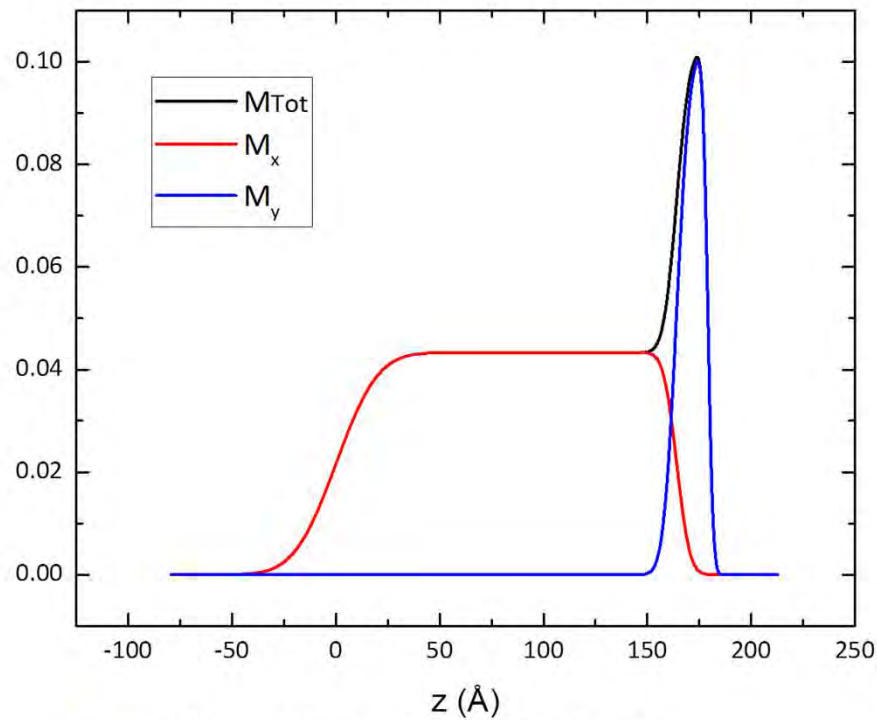
ISIS

Orthogonal YIG

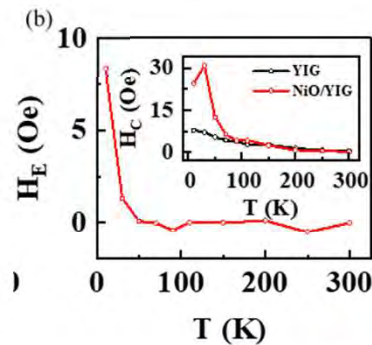


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ISIS



- Pt (4 nm)/NiO (2 nm)/YIG/GGG
- Significant Spin flip scattering
- NiO Quantization axis orthogonal to YIG
- Uncompensated moment in NiO
- Domain state model
- EB at low temperature



Nowak, U. et al. Phys. Rev. B **66**, 014430 (2002)



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ISIS

Small Angle Scattering

- Grazing Incidence to give depth selectivity
- Difference gives the interference term

$$I^+(Q, \alpha) = \langle |F^{++}|^2 \rangle + \langle |F^{+-}|^2 \rangle \\ = F_N^2 + (F_M^2 - 2PF_NF_M) \sin^2 \alpha$$

$$I^-(Q, \alpha) = \langle |F^{--}|^2 \rangle + \langle |F^{-+}|^2 \rangle \\ = F_N^2 + (F_M^2 + 2P\epsilon F_NF_M) \sin^2 \alpha$$

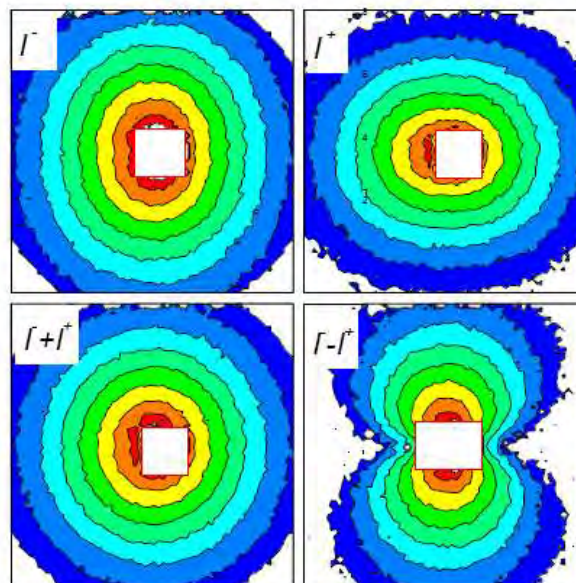


Figure 1
SANSPOLE patterns in Fe_3O_4 for neutron spins antiparallel (I^-) and parallel (I^+) to the horizontal field. The arithmetic mean $[I^- + I^+]/2$ corresponds to the 2D pattern of non-polarised neutrons. The difference $(I^- - I^+)$ yields the interference term [equation (1c)].

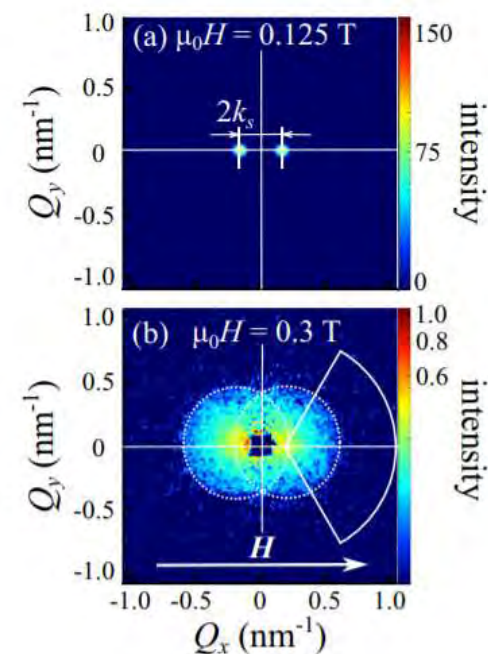


FIG. 1. Maps of the small-angle neutron scattering at $T = 15$ K for $\text{Fe}_{0.7}\text{Co}_{0.3}\text{Si}$: (a) in the conical phase at magnetic field $\mu_0H = 0.125$ T, (b) in the induced ferromagnetic phase at magnetic field $\mu_0H = 0.3$ T.

- A. Wiedenmann J. Appl. Cryst. (2000). 33, 428-432
- Grigoriev et al. Phys Rev B **100**, 094409 (2019)
- Mühlbauer, S. et al. Magnetic small-angle neutron scattering. Rev. Mod. Phys. **91**, 015004 (2019).



Summary & Outlook

- The neutron possess a range of characteristics well matched to both *curiosity driven* and *technologically* relevant nanoscale research
 - Absolute, quantitative results provide a robust test of theory
 - From atomic and nano through to micro-macroscopic lengthscales
- Examples drawn from:
 - Ferromagnetic Insulators
 - Chiral Magnetism
 - Magnetocaloric
 - *Biomedical applications, Clean Energy*
- Frontier Research
 - Thin film quantum matter
 - Novel sample environment
 - Smaller Samples
 - Increasing connection with calculation/theory

